Effect of potassium application in forest soil on ^{137}Cs levels in plants and fungi

K. Rosén[1,*], M. Vinichuk[1,2],T. Nilsson[1], I. Nikolova[1], K. Johanson[1],
[1]*Department of Soil and Environment, Swedish University of Agricultural Sciences, SLU, P.O. Box 7014 SE-750 07, Uppsala, Sweden*
[2]*Department of Ecology, Zhytomyr State Technological University, 103 Chernyakhovsky Str., 10005, Zhytomyr, Ukraine*
** Corresponding author. Tel.: +46 018 67 12 85; fax: +46 018 67 28 95.*
E-mail: Klas.Rosen@slu.se (K. Rosén)

Abstract. Low-growing perennial shrubs, heather (*Calluna vulgaris*), lingonberry (*Vaccinium vitis-idaea*) and bilberry (*Vaccinium myrtillus*) as well as fungal species growing on K-fertilized plots (100 kg K ha^{-1} has been applied in 1992) showed significantly lower ^{137}Cs activity concentrations than corresponding species growing in a non-fertilized control area. ^{137}Cs activity concentration in both plants and fungal sporocarps sampled on K-fertilized area decreased over years but was as an average up to 60% lower compared to ^{137}Cs activity levels in the same species sampled on control area. The reduction of ^{137}Cs activity concentration due to fertilization of forest soil with K was found to be long lasting, statistically significant and strongly pronounced in all species studied. Notable decrease of ^{137}Cs activity concentrations in heather, lingonberry and bilberry was observed already within the first year after K fertilizer application, however, the most pronounced and significant reduction occurred over the first 7-8 years, followed by a gradually decreasing effect. The reduction in ^{137}Cs activity concentration in fungi after K fertilization was less pronounced, but still statistically significant. It has been suggested that single application of K fertilizer to forests might be an effective and feasible long-term countermeasure to decrease radiocaesium accumulation by plants and fungi. The data is published in Journal of Environmental Radioactivity 102 (2011) 178-184.

1. INTRODUCTION

Radiocaesium (^{137}Cs) uptake by plants and fungi after Chernobyl in boreal forest ecosystems in Scandinavia has been very high. In the following years activity concentrations of ^{137}Cs in forest plants, fungal sporocarps and game animals remained relatively high and decreased only slowly [1]. In edible fungi species ^{137}Cs activity concentration either did not decrease over the last 20 years (*Suillus variegatus*) or even significantly increased (*Cantharellus* spp.) [2]. Thus, combination of high ^{137}Cs activity concentrations in forest food products and the long effective ecological half-life of ^{137}Cs in forest ecosystems can result in high time-integrated intake of ^{137}Cs by humans through local non-commercial consumption of berries, fungi, moose and roe deer.

^{137}Cs uptake by plant species, particularly those belonging to the Ericaceae family such as heather (*Calluna vulgaris*), has decreased over the past 15-20 years but is still rather high, compared to other vascular species, such as cloudberry (*Rubus chamaemorus*) and cranberry (*Vaccinium oxycoccus* and *Vaccinium microcarpon*) [3]. The ^{137}Cs levels in fungal species that are less prone to accumulating ^{137}Cs can be similar to the levels found in bilberry and lingonberry, but most fungal species can have 10-100 times higher levels than the vascular plant species. Potassium (K) fertilization has been shown to decrease soil-plant transfer of ^{137}Cs in agricultural ecosystems [4]. The effect of K fertilization in reducing ^{137}Cs uptake by forest plants growing on radionuclide-polluted soils has been studied in a few experiments, e.g. using Scots pine trees [5], lingonberries [6] and shoots of heather [7]. No studies have been performed

to date on the long-term effects of K application on ^{137}Cs uptake by fungi. The aim of fertilizing forests contaminated by long-lived ^{137}Cs with K-containing products is to achieve a long-term reduction in the activity concentration of ^{137}Cs in plants and fungi and to reduce its transfer in the food web. The aim of the present study was to investigate the long-term effects of a single K fertilizer addition on ^{137}Cs uptake by heather, lingonberry, bilberry and fungi over the period 1992-2009. Our hypothesis was that K fertilization would bring about a long-term reduction in ^{137}Cs levels in plants and fungi in boreal forest ecosystems.

2. MATERIALS AND METHODS

2.1. Study area

The study forest area is located in Harbo (Heby commune), about 40 km north-west of Uppsala in central Sweden. The ground deposition of ^{137}Cs in 1986 was 35-40 kBq m^{-2} [8]. A control area (K-) of about 1.0 ha was chosen in a rocky part in the boreal forest (N 66°59'50″; E 15°73'60″). The dominating soil types within the area are Folic Leptosoil and Haplic Podzol developed on sandy glacial till with mor humus type [9] with organic matter content in upper organic reach layer more than 70% and pH 3.4-3.7. Ammonium acetate lactate extractable K varied between 480 (upper organic layer) to 35 (mineral horizon) mg kg^{-1}. The forest is dominated by Scots pine (*Pinus sylvestris*) and Norway spruce (*Picea abies*). In the field layer, bilberry, lingonberry and heather are the predominant dwarf shrub species. A similar rocky area of about 1.0 ha in the immediate vicinity of the control area (N 66°59'50″; E 15°74'60″) was chosen for the K fertilization treatment, which was applied to three specific plots (area K+).

2.2. Fertilization

Potassium chloride (KCl) was spread once as a single dose in the beginning of June 1992 using a conventional centrifugal broadcasting spreader commonly used in agriculture. The amount of applied potassium virtually corresponded to about 30-40 % of available pool of K in soil.

2.3. Sampling

Sampling of aboveground green parts of heather, lingonberry and bilberry was carried out before (May) and after K fertilizer application (August and September) in 1992. In the following years (1993-2009), samples were collected on one occasion per year, usually in August-September.

2.4. Radiometry and statistical analyses

^{137}Cs in plants and fungi samples were determined using well-calibrated HPGe detectors at the Swedish University of Agricultural Sciences (SLU), Uppsala. All activity concentrations are expressed as Bq kg^{-1} dry weight (d.w.) and decay corrected to the sampling date. Prior to analyses, data were checked for normality using the Anderson-Darling normality test and $\log_{10}$-transformed in order to achieve normality when necessary. A non-parametric Mann-Whitney test was performed if necessary to detect difference between variances. All

statistical analyses were carried out using Minitab (© 2007 Minitab Inc.) software.

3. **RESULTS**

3.1. Plants

Mean values of ^{137}Cs activity concentrations in heather, bilberry and lingonberry sampled over the period 1992-2009 are shown in Figure 1 and Table 1. The effect of K fertilization on ^{137}Cs uptake in heather was most pronounced over the first nine years (1992-2000) after fertilization and became smaller but still significant during recent years (2001-2009). A similar trend was observed for bilberry, with a greater reduction effect during the first eight years (1993-2000) and a smaller effect in the last years of the study (2000-2009). In the case of lingonberry the effect of K application seemed to be more long-lasting, but with the difference in ^{137}Cs activity concentrations in plants grown on fertilized and control areas also gradually decreasing over recent years. An increase in ^{137}Cs activity concentration in plants occurred in the periods 1997-1998 and 2000-2001 on both K-fertilized and control areas (Figure 1).

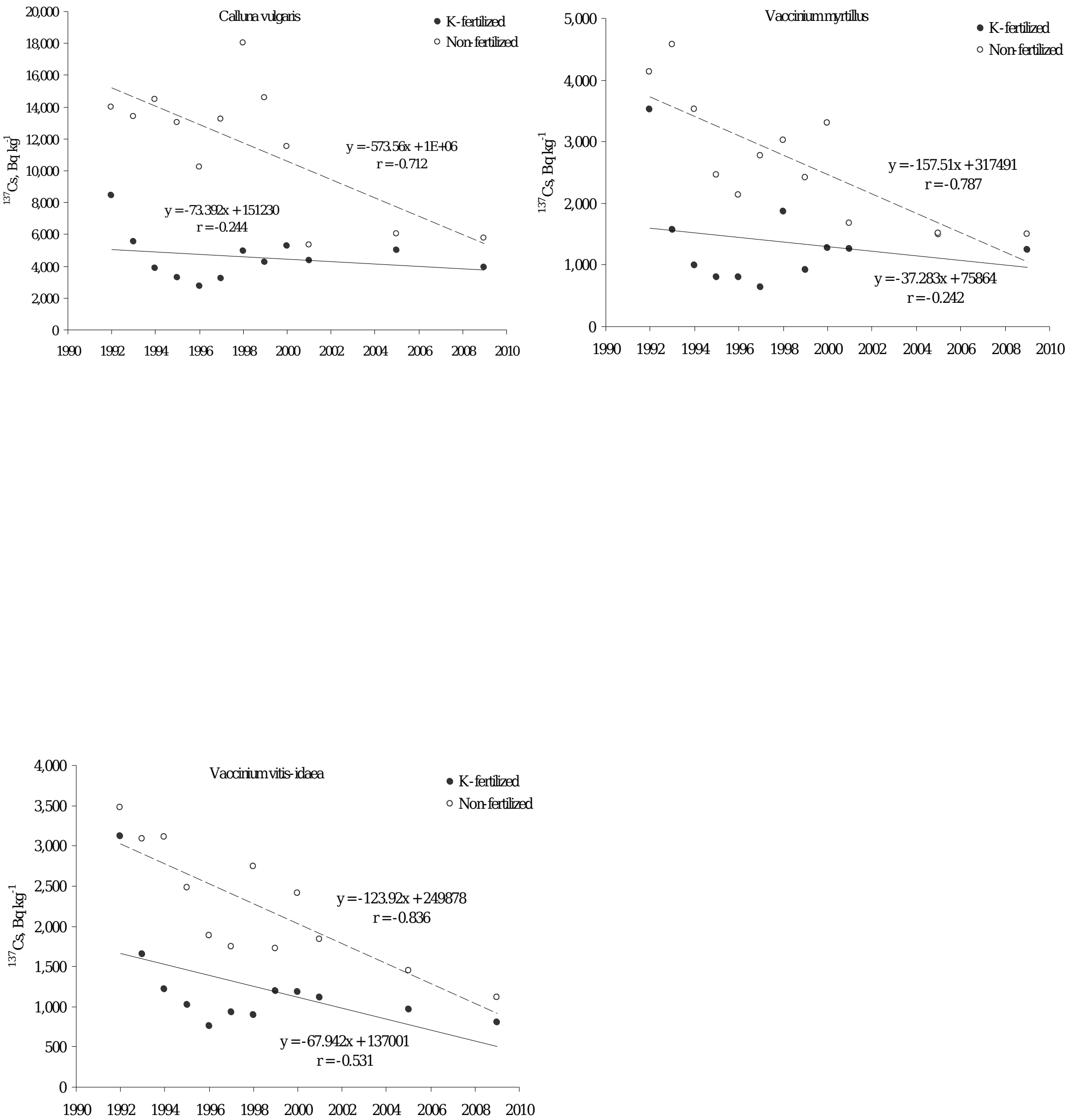

Figure 1. ^{137}Cs activity concentrations in heather (*Calluna vulgaris*), bilberry (*Vaccinium myrtillus*) and lingonberry (*Vaccinium vitis-idaea*) as a function of time on K-fertilized (K+, solid line) and non-fertilized (K-, dotted line) areas.

Regression analyses evidences that all three plant species studied showed similar decreasing trends of ^{137}Cs activity concentration following application of K fertilizer. The decrease in ^{137}Cs activity concentration in plants grown on the control area was also rather similar but more pronounced. The mean ^{137}Cs activity concentrations on the K-fertilized area and on the control area were in the range 4 560-11 580 Bq kg^{-1} for heather), 1 410-2 690 Bq kg^{-1} for bilberry and 1 300-2 160 Bq kg^{-1} for lingonberry. Plants of all three species grown on the K-fertilized area showed a highly significant ($p < 0.001$) decrease in ^{137}Cs activity of 40-60% compared with plants grown on the control area (Table 1).

Table 1. Mean 137Cs activity concentration (Bq kg-1) in plants collected from K-fertilized (K+) and non-fertilized (K-) areas over the period 1992-2009.

Species	n	K-	K+	K+/K-, %[1]
Calluna vulgaris	89	11 584	4 563	60.6***
Vaccinium myrtillus	86	2 691	1 409	47.6***
Vaccinium vitis-idaea	90	2 162	1 295	40.1***

[1] ((^{137}Cs in plants from K- area - ^{137}Cs in plants from K+ area) / ^{137}Cs in plants from K- area)) x 100; *** $p < 0.001$;

There was a distinct difference in ^{137}Cs activity concentration between K-fertilized and non-fertilized areas for all three plant species and the effect varied over time. It was less pronounced in the year of fertilization (1992) and by the end of the study period (2001-2009), while the effect of fertilization was very clear in the period 1993-2000. After K fertilization the system came to a ‘steady state’ which was maintained for about 7 years, during which the difference in ^{137}Cs activity concentration compared with the control area was at its highest: 53% for lingonberry, 63% for bilberry and 69% for heather. By the end of the experiment (2001-2009) this difference had decreased to 34, 15 and 22% respectively. Assuming an exponential decrease in ^{137}Cs activity concentrations due to radioactive decay in plants collected on non-fertilized areas, the long-term rate of change in ^{137}Cs concentration can be described in terms of an effective ecological half-life, T_{eff} (years), where $T_{eff} = \ln(2)/\lambda$ [10]. The ecological half-life of ^{137}Cs (time taken for a given activity to decrease to half the original value) for plants growing on the control area over the 17-year sampling period was found to be about 11 years for bilberry and lingonberry and about 9 years for heather. These estimates fit well with data presented by Pröhl et al., [11], who reported an ecological half-life for ^{137}Cs in bilberry leaves of 9.2 years in spruce forest and 11 years in forest and peat bog. The long-term rate of change in ^{137}Cs activity concentrations in plants growing on the fertilized area in terms of T_{eff} is a “double exponential” due to the effect of K application (i.e. T_{eff} increases over time).

3.2. Fungi

Species of *Suillus variegatus, Lactarius rufus*, *Cortinarius semisanguineus* and *Rozites caperata* produced sporocarps nearly every year and were collected in most years of the study on both K-fertilized and control areas. *Suillus variegatus* and *C. semisanguineus* both showed a distinct peak in ^{137}Cs activity concentration in 1998 (Figure 2). The inhibiting effect of K application on ^{137}Cs uptake was observed in *S. variegatus* over a period of 10 years, (except 1998). In sporocarps of *C. semisanguineus* the effect of K was apparent in 6 years out of the 8 for which data were available. A similar decrease in ^{137}Cs activity concentration was detected in *L. rufus* (in 6 years out of 7). In sporocarps of *R. caperata*, a reduction effect was apparent in the 4 years when comparisons were possible.

Regression analyses were employed to determine the statistical relationship between ^{137}Cs activity concentrations in sporocarps collected on K-fertilized and non-fertilized areas. It is apparent from the diagram that sporocarps of *S. variegatus, C. semisanguineus* and *L. rufus* collected on the control area showed a similar gradual decrease in ^{137}Cs activity concentrations, whereas the decrease in ^{137}Cs activity concentration in sporocarps of *R. caperata* was more pronounced. The pattern of ^{137}Cs activity concentration decrease in sporocarps collected on K-fertilized plots was also rather similar for *S. variegatus* and *C. semisanguineus* but slightly different for sporocarps of *L. rufus* and *R. caperata*.

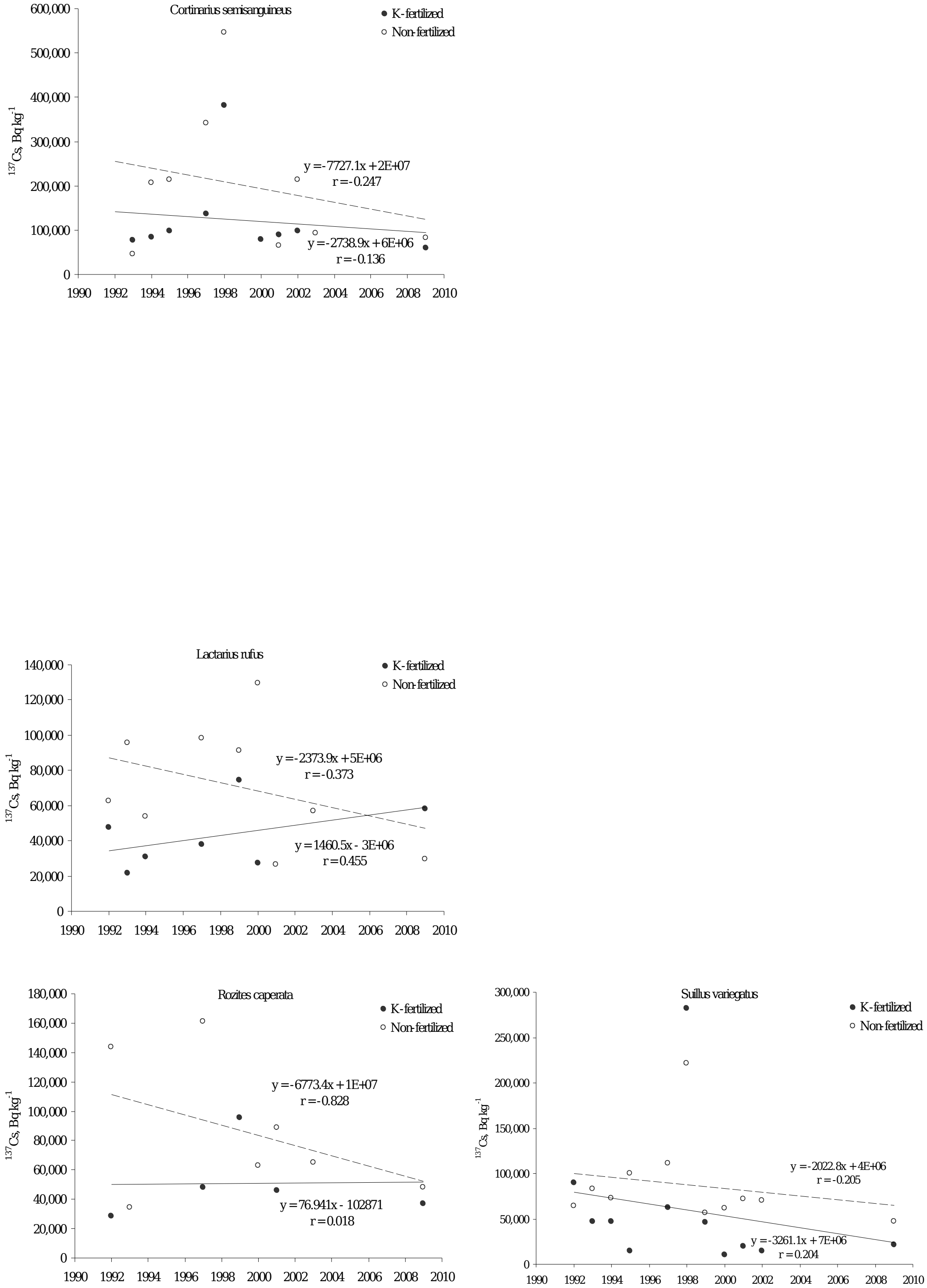

Figure 2. ^{137}Cs activity concentrations (Bq kg^{-1}) in fungal sporocarps as a function of time on K-fertilized (K+, solid line) and non-fertilized (K-, dotted line) areas.

Mean ^{137}Cs activity concentrations in fungal sporocarps collected over the period 1992-2009 are presented in Table 2. The ^{137}Cs activity concentrations in all four species of fungi grown on the K-fertilized area were lower (with a range 34 300-134 110 Bq kg^{-1}) than those in the corresponding species collected on the control area (with a range 81 980-221 850 Bq kg^{-1}). Potassium application resulted in a 21-58 % decrease in ^{137}Cs activity in the species studied. All these differences were statistically significant ($p < 0.05$).

Table 2. Mean ^{137}Cs activity concentration (Bq kg^{-1}) in fungi collected from K-fertilized (K+) and non-fertilized (K-) areas over the period 1992-2009.

Species	n	K-	K+	K+/K-, %[1]
Cortinarius semisanguineus	65	221 853	134 112	39.5*
Lactarius rufus	22	81 978	34 297	58.2***
Rozites caperata	17	91 877	50 975	44.5*
Suillus variegatus	62	86 286	68 401	20.7***

[1] ((^{137}Cs in fungi from K- area - ^{137}Cs in fungi from K+ area) / ^{137}Cs in fungi from K- area)) x 100; *** $p < 0.001$; * $p < 0.05$

The frequency of appearance of fungal sporocarps varied greatly from year to year, making it more difficult to determine the effects of K fertilization. Of the fungal species sampled, *Lactarius rufus* showed a similar reduction in ^{137}Cs activity concentration after K application as vascular plants – about 58%. The other fungal species showed smaller differences compared with the control area but still had significantly reduced ^{137}Cs uptake when grown on K-fertilized plots. Overall, the decrease in ^{137}Cs activity concentration in fungi due to K fertilization was less consistent and less pronounced than in vascular plants, which was partly due to the ^{137}Cs activity concentration in fungal sporocarps varying significantly within the study period. There also appeared to be a variation in long-term ^{137}Cs activity concentration trends among the four species of fungi studied, which is difficult to explain, but might be due to the complexity of the interactions between fungi and environment (hosts). In a long-term study in the same area of Sweden by Mascanzoni [2], ^{137}Cs uptake by *Suillus variegatus* did not decrease over the period 1986-2007, while uptake by *Cantharellus* spp. even exhibited a significant increase. The ambiguous response of fungi to K fertilization might be due to the relationships between K and Cs when taken up by fungi, which are not well understood and it has been suggested that the mechanism of Cs uptake by fungi might be completely different from that of K uptake [12], [13]. Nevertheless, the effect of K application to forest seemed to persist for more than one decade. Even after 17 years, there were quite substantial decreases in the ^{137}Cs activities in dwarf shrubs and wild fungi growing in the fertilized area. Similarly, a long-lasting effect of K-containing fertilizers on caesium uptake in different Scots pine compartments was reported by Kaunisto et al. [5].

Thus, a single K fertilizer application was able to reduce ^{137}Cs uptake by forest plants and fungi over a period up to 17 years. It is well known from studies on farmland that K fertilization can be an efficient method to reduce crop uptake of radiocaesium and indeed K fertilization was used as a remedial measure in Swedish agricultural systems in 1986 and 1987 [4]. The most effective K dose on grassland was reported to be 200 kg K ha^{-1}, giving the highest recorded reduction in total caesium transfer of about 60-80%, when application of 100 kg K ha^{-1} reduced ^{137}Cs uptake by 60-65%. On the basis of the current results, K application to forest ecosystems seems to be equally effective. The most pronounced effect in the vascular plant species studied was observed

in heather, which demonstrated a 60% reduction. The decrease in ^{137}Cs activity concentration in plants was found to be more consistent than in fungi within the study period.

The reason for the long-term effects observed in the different species of plants and fungi is not fully understood, but it seems reasonable to assume that they may be due to continuing competition between ^{137}Cs and K^+ ions in the soil. As long as K^+ is available in the soil, its uptake by plants presumably increases, which in turn results in a decrease in ^{137}Cs activity concentration in plants. The gradual decline in the effects of K application on ^{137}Cs uptake by plants in subsequent years might be due to K is also biologically bound to other forest species over time, such as trees. It is also plausible that significant reduction in plant and fungi uptake of ^{137}Cs after K application is due to low K status in boreal forest soils.

4. Conclusions

A single application of K fertilizer at a rate of 100 kg K ha^{-1} to boreal forest is an effective way to decrease ^{137}Cs activity concentrations in plants and fungi over a period of more than one decade. Decrease of ^{137}Cs activity concentrations in heather, lingonberry and bilberry was observed even within the first year. The most pronounced and significant reduction occurred over the first 7-8 years, followed by a gradually decreasing effect. The reduction in ^{137}Cs activity concentration in fungi after K fertilization was less pronounced, but still statistically significant.

Acknowledgements

The data in this manuscript is published in 14. We would like to express our thanks to editor of J. Environ. Radioactivity and to MSc. T. Nilsson for assistance with experiments and to Dr. A. Dahlberg for assistance with identification of fungal species. The project was financially supported by SKB (Swedish Nuclear Fuel and Waste Management Co) and the Swedish University of Agricultural Sciences (SLU).

References

1. R.T. Palo, N. White, K. Danell, Wildlife Biol. 9 (2003) 207-212.
2. D. Mascanzoni, J. Radioanal. Nucl. Ch. 282 (2009) 427-431.
3. K. Rosén, M. Vinichuk, K. Johanson, J. Environ. Radioactiv. 100 (2009) 534-539.
4. K. Rosén, PhD. thesis. (1996)
5. S. Kaunisto, L. Aro, A. Rantavaara, Environ. Pollut. 117 (2002) 111-119.
6. T. Levula, A. Saarsalmi, A. Rantavaara, Forest Ecol. Manag. 126 (2000) 269-279.
7. M. Strandberg, M. Johansson, J. Environ. Radioactiv. 40(2) (1998) 175-184.
8. SGAB, Uppsala, (1986).
9. IUSS. World Reference Base for Soil Resources 2006. No. 103. (FAO, Rome, 2007) pp. 55-68.
10. J.T. Smith, S.V. Fesenko, B.J. Howard, A.D. Horril, N.I. Sanzharova, et al., Environ. Sci. Technol. 33 (1999) 49-54.
11. G. Pröhl, S. Ehlken, I. Fiedler, G. Kirchner, E. Klemt, et al., J. Environ. Radioactiv. 91 (2006) 41-72.
12. S. Yoshida, Y. Muramatsu, J. Environ. Radioact. 41 (1998) 183-205.

13. M. Vinichuk, A.F.S. Taylor, K. Rosén, K. Johanson, Sci. Total Environ. 408 (2010) 2543–2548.
14. K. Rosén, M. Vinichuk, I. Nikolova, K.J. Johanson J. Environ. Radioactiv. 102 (2011) 178-184.